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ABSTRACT

We present collective Thomson scattering with soft x-ray free electron laser radiation as a method to track the evolution of warm dense matter plasmas with ~200 fs time resolution. In a pump-probe scheme an 800 nm laser heats a 20 µm hydrogen droplet to the plasma state. After a variable time delay in the order of ps the plasma is probed by an x-ray ultra violet (XUV) pulse which scatters from the target and is recorded spectrally. Alternatively, in a self-Thomson scattering experiment, a single XUV pulse heats the target while a portion of its photons are being scattered probing the target. From such inelastic x-ray scattering spectra free electron temperature and density can be inferred giving insight on relaxation time scales in plasmas as well as the equation of state. We prove the feasibility of this method in the XUV range utilizing the free electron laser facility in Hamburg, FLASH. We recorded Thomson scattering spectra for hydrogen plasma, both in the self-scattering and in the pump-probe mode using optical laser heating.

Keywords: inelastic x-ray scattering, Thomson scattering, warm dense matter, free electron laser

1. INTRODUCTION

1.1 Warm dense matter – challenges and opportunities

Warm Dense Matter (WDM) is a challenge to modern physics. It is a plasma state with free electron temperatures T_e of some eV and free electron densities n_e=10^{21}-10^{26} cm^{-3}, which are near and beyond solid density. Therefore, it is an intermediate state between condensed matter and the ideal plasma. Significant progress has been made in describing the physical properties of condensed matter as well as ideal, hot plasmas. Nevertheless, the intermediate regime, i.e. WDM, combines the challenges from both fields, namely strong coupling of the particles and thermally determined plasma dynamics. This is one reason why WDM is vastly unknown. Another one is the difficulty to generate homogeneous quantities larger than µm^3 as optical lasers, for example, have nm penetration depths in solids leading to strong gradients. On top of that, laboratory WDM tends to expand, fragment or collapse at the latest within ns into ideal plasmas or condensed matter.

However, these challenges are worthwhile undertaking because WDM is present as a transient state in many laser matter experiments and even statically in planet interiors [1, 2] where it is gravitationally confined. WDM is thus of great interest in the context of laser matter interaction, in particular inertial confinement fusion [3], and also for astrophysics [4, 5].

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Another opportunity lies in the common principles that WDM shares with other coupled systems such as semiconductors and high temperature superconductive materials. They also require the understanding of many body particle correlations under strong interaction. Hence, synergy effects from a general understanding of these systems can be expected. Finally, high impact applications and spin offs are anticipated, e.g. in the energy, health, telecommunications and chip sector [6].

We generate and investigate warm dense hydrogen. This is possible by exciting liquid hydrogen with an intense and ultra fast x-ray ultra violet (XUV) photon pulse and recording the scattered photons from the same pulse (self-Thomson scattering). On top of that, the investigation of relaxation time scales is possible in a pump probe scheme with a ps delayed XUV pump and probe pulse or an optical pump and XUV probe pulse. In all cases, the scattered XUV photons are recorded spectrally. These inelastic x-ray scattering spectra are interpreted to determine the main plasma parameter, free electron density and temperature.

1.2 Inelastic x-ray scattering and collective Thomson scattering

Inelastic x-ray scattering (IXS) is meanwhile a well developed technique to probe plasmas and in particular their free electron density, free electron temperature and degree of ionization. Optical laser based inelastic scattering was initially used to diagnose theta- and z-pinch as well as magnetically confined plasmas [7, 8]. These plasmas had low enough free electron densities so that optical photons could penetrate and hence probe the plasma. For that to be the case, the photon frequency ω_{light} needs to be higher than the electronic plasma frequency ω_{pe},

$$\omega_{light} > \omega_{pe} = \sqrt{\frac{n_e e^2}{\varepsilon_0 m_e}} ,$$

where e is the elementary charge, ε_0 the vacuum permittivity, and m_e the electron rest mass. Figure 1 depicts this relationship between plasma frequency and free electron density. Consequently, if high density plasmas, in particular WDM, are to be probed via IXS, high photon frequencies or short wavelengths have to be used in order to penetrate and probe through the critical density surface. The transfer of this technique into the x-ray regime has been demonstrated [9-13] where optical laser generated plasmas are used as sources for x-ray probe radiation.

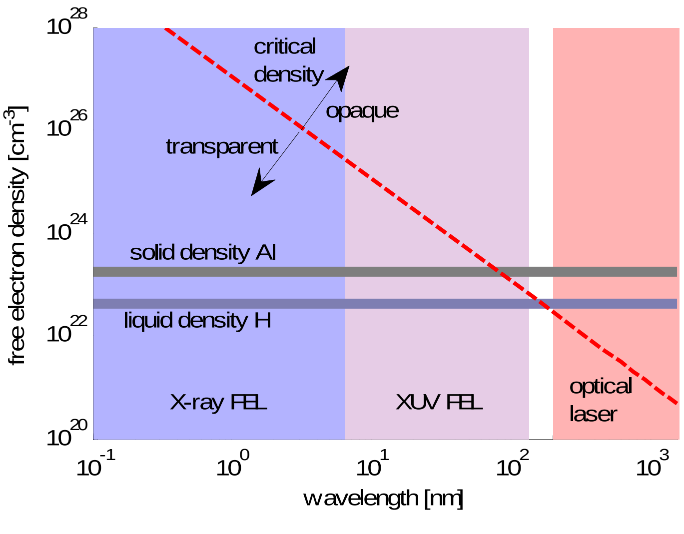


Fig. 1. (color online) Critical free electron density over wavelength (red, dashed) in comparison to hydrogen and aluminum solid density and regimes of some current and future light sources. Free electron laser sources like the European XFEL and FLASH are capable of probing higher than solid density plasmas.

In particular, we are interested in the collective electron motion which is described by collective Thomson scattering. Therefore, we record inelastic scattering spectra with red and blue shifted peaks with respect to the incident radiation, the plasmons. The photon energy shift of the plasmons is symmetric to the incident radiation and given in the long wavelength limit by ω_{res} [14],

$$\omega_{res}^2 = \omega_{pe}^2 + \frac{3k_B T_e}{m_e} k^2 ,$$

where k_B is the Boltzmann constant and $k = 4\pi\lambda_0^{-1} \sin(\Theta/2)$ the wave vector difference between incident and scattered light for small photon energy transfers and plasma frequencies in comparison to the photon frequency. The incident wavelength is λ_0 and the scattering angle Θ. Furthermore, the spectral dependence of Thomson scattering is described by the structure factor $S(k,\omega)$ which exhibits a ratio between red shifted amplitude $S(-k,-\omega)$ and blue shifted amplitude $S(k,\omega)$ determined by the detailed balance relationship,

$$\frac{S(-k,-\omega)}{S(k,\omega)} = e^{-\hbar\omega/k_B T_e} .$$

For this measurement, the electrons in the sample are required to be thermally distributed throughout the scattering process. As a result, if one can resolve the plasmons in a collective Thomson scattering spectrum one knows the plasma temperature via the plasmon intensity ratio and simultaneously the free electron density via the photon energy shift. Therefore, collective Thomson scattering is very attractive for the investigation of plasmas [15-17]. A schematic spectrum is depicted in figure 2.

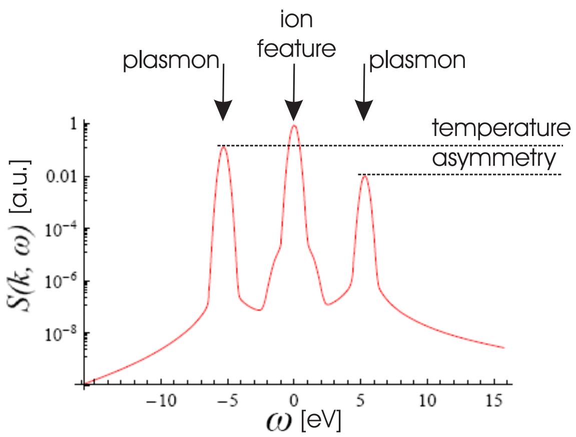


Fig. 2. (color online) This simulation shows a typical collective Thomson scattering spectrum over the change in photon energy with the elastically scattered ion feature and the red and blue shifted plasmon peaks.

1.3 Plasma physics with x-ray free electron lasers

Due to the small cross section of Thomson scattering, $0.665 \cdot 10^{-24}\,\mathrm{cm}^2$, one needs a source of probe light that can deliver many photons on the sample. On top of that, the time resolution of a pump probe experiment is limited by the pulse duration of pump and probe as well as the temporal jitter between them. The pulse duration as well as the amount of photons deliverable onto the target can be expressed in terms of peak brilliance of a source, i.e. the emitted photon number per time, emission angle, source size and 0.1% bandwidth interval. One also needs higher photon energies to penetrate and probe higher plasma densities. Figure 3 depicts the key quantities, peak brilliance and photon energy, for several current and future light sources. Here, one can immediately see that free electron laser (FEL) sources, such as the free electron laser in Hamburg, FLASH, as well as the European XFEL and the Linac Coherent Light Source, LCLS, are ideal for the investigation of dense plasmas using IXS. That is because they combine high photon energies with high peak brilliance. Hence, XUV and x-ray FEL sources are of general interest for plasma physics applications [18, 19] and have also yielded first novel results [20, 21].

We demonstrate and utilize collective Thomson scattering in the XUV regime where the probe radiation is provided by FLASH [22, 23]. Here, the light pulses are delivered with 5 Hz repetition rate which allows us to integrate over several probe pulses if necessary.

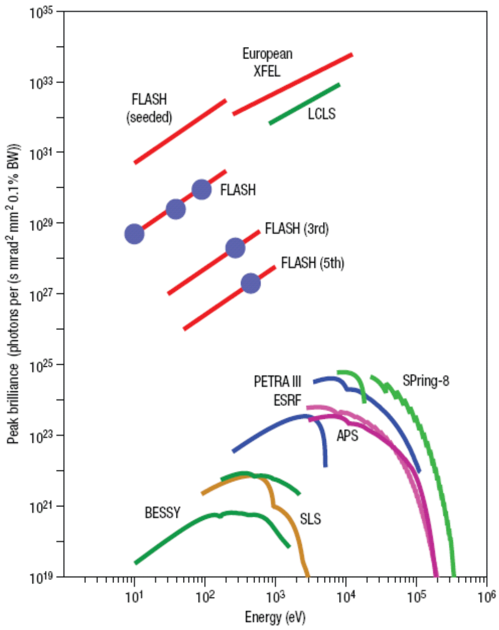


Fig. 3. (color online) Peak brilliance of current and future light sources over photon energy (from [22]).

2. EXPERIMENTAL SETUP

Figure 4 depicts a schematic of the experimental setup. A liquid hydrogen target with 20 µm diameter is injected horizontally into the vacuum chamber and drifts to the point of interaction. Synchronized to its arrival and during its ballistic motion it is irradiated by a pump beam which can be part of the FEL beam itself or an 800 nm laser transforming it into the WDM state. Irradiation with the FEL (13.5 nm, ~40 fs, ~3×10^{14} W/cm^2) heats the target homogeneously and isochorically due to its high penetration depth (9.4 µm [24]) and short pulse duration. The exact FEL pulse energies are measured parasitically with a gas monitor detector [25]. On the other hand, pumping via the 800 nm laser (100 fs, 3×10^{15} W/cm^2) will generate a highly ionized surface which can reflect a major part of the pulse as soon as the critical density is reached. Nevertheless, the bulk of the target can still be heated via hot electron production at the critical surface [26, 27]. Both pump options are available at FLASH. In either case, the pump pulse is followed by an FEL probe pulse with variable delay in the range of 0 ps to 100 ps. The jitter between an optical 800 nm laser and the FEL can be measured and thus reduced to an accuracy of as small as 200 fs [28] for single pulses and <500 fs for longer integrations. Experiments which split a single FEL pulse into a variably delayed pump and probe achieve even higher accuracies [29, 30]. Either way, the IXS signal from the FEL probe pulse is recorded spectrally. This setup is capable of tracking the temporal evolution of warm dense hydrogen plasma with the ultimate goal of gaining insight into the WDM equation of state. Also, self-Thomson scattering experiments can be realized with a single FEL pulse. In this scheme a single pulse heats the sample while the scattering from the same pulse is recorded. Thus, we gain information about the plasma on time scales of the FEL pulse duration (40 fs) and about the influence of the probe pulse itself in a pump-probe scheme. More details on the setup can be found in [31, 32]. Simulations on the FEL matter interaction, target homogeneity and impact on the scattering spectrum have also been conducted [33]. They indicate that plasmons can still be observed even when slight gradients are present in the plasma distribution. This inhomogeneity would lead to a broadening of the plasmons. In general, plasmon spectra are fairly robust to such disturbances and even non-equilibrium [34].

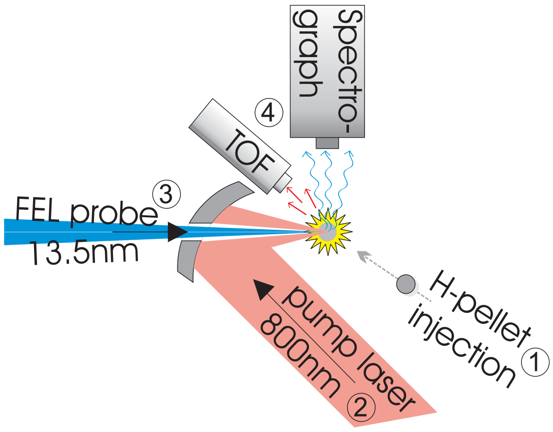
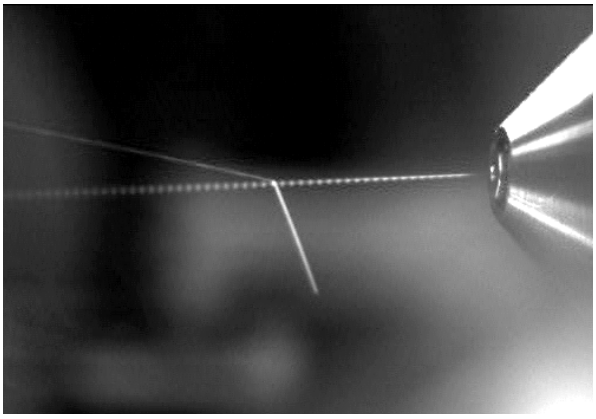


Fig. 4. (color online) Schematic of the experimental setup with an 800 nm pump and 13.5 nm FEL probe beam. The H-pellet is injected into the interaction region (1), hit by the pump laser (2), probed by the variably delayed FEL beam (3) while the scattering spectrum is recorded (4). A field free electron time of flight (TOF) spectrometer can give further information on how well the hydrogen droplet was hit by pump and probe pulse. A continuous jet of liquid hydrogen can also be produced by switching off the piezoelectric actuators.

2.1 The hydrogen droplet source

In order to make use of the 5 Hz repetition rate of the FEL by integrating scattering spectra over several pulses one also needs to provide a target with sufficient refresh rate. We utilize a liquid hydrogen jet. The source of this jet cools the hydrogen by liquid helium to around 20 K and injects it into the vacuum chamber through a 20 µm nozzle with about 1 bar pressure. The hydrogen reaches a speed of around 60 m/s which guarantees a new target for every FEL pulse. Also, hydrogen has with Z=1 the lowest number of bound electrons and, therefore, a small elastic scattering signal. This facilitates to spectrally resolve the plasmon peaks from the elastically scattered signal.

Moreover, the nozzle is equipped with piezoelectric actuators which can induce a Rayleigh break up in the liquid hydrogen jet causing the reproducible formation of droplets. The size and separation of the droplets can be adjusted via the vibration frequency of the actuators. The still image of video 1 shows the nozzle ejecting a modulated hydrogen jet. The corresponding video shows how the modulation of the hydrogen jet can be tuned. By changing the phase of the actuator frequency with respect to the FEL pulse one can change the location of the droplets during that event. This way, one can synchronize the arrival of a droplet at the interaction point with the arrival of an FEL pulse. After switching off the piezoelectric actuators the string of droplets transforms back into a continuous jet.



Video 1. Modulated hydrogen jet ejected from the cryogenic nozzle into the vacuum chamber. In this particular frame one can see how a droplet collides with another droplet which has been reflected from the chamber walls and is not synchronized anymore to the stroboscopic camera. The video shows the tuning of the hydrogen jet modulation via the frequency at which the piezoelectric actuators are driven. By changing the frequency one can set size and spacing of the droplets. http://dx.doi.org/doi.number.goes.here

2.2 The high throughput and resolution spectrograph for xuv light (HiTRaX)

In order to acquire and resolve the Thomson scattered signal a special spectrograph for XUV light was constructed. Several requirements have to be met by this instrument, which is the main diagnostic. It needs to have sufficient resolution to distinguish the plasmons from the elastically scattered signal between which a separation of about 1 eV at around 92 eV (13.5 nm) photon energy can be expected. A large acceptance solid angle and efficiency of the optical components is crucial in order to compensate for the small Thomson scattering cross section. It has to incorporate filters to suppress visible light, has to be sufficiently small to be mounted on top of the vacuum chamber and easy to align. All these requirements are met by the high throughput and resolution spectrograph for XUV light (HiTRaX). Figure 6 shows the instrument. For alignment purposes it is mounted inside a CF100 x-y-z-vacuum manipulator. The filter wheel contains up to seven filters and is motorized. Also motorized are the grating and debris protecting shutter which, altogether, make the spectrometer a versatile tool. The high collection solid angle ($1.9\cdot10^{-3}\,\mathrm{sr}$) is achieved by a toroidal mirror and the dispersion by a variable line space grating, both operated at grazing incidence angle in order to optimize reflectivity for the XUV radiation. This results in a throughput, i.e. counts per photon into 4 π, as high as $3.5\cdot10^{-4}$ at 13.5 nm and a resolution of $\lambda/\Delta\lambda = 250$. Details on this instrument will be described in a forthcoming publication.

Fig. 5. (color online) Picture of the high throughput and resolution spectrograph for XUV light (HiTRaX), which is the main diagnostic for the Thomson scattered radiation. It is composed of a toroidal collection optic, a variable line space grating, rotatable filter wheel, a shutter to protect from debris and housed within the cut open CF100 x-y-z-manipulator.

3. EXPERIMENTAL RESULTS

During the experiment we had to face several challenges. One of them was the vacuum requirements at the beam line. In order to connect the experimental chamber with the light source 2500 l/s turbo pumping power and several differential pumping stages were necessary to sustain the chamber pressure of ~10^{-5} mbar while the hydrogen source was in operation. Also, an exceptional performance of the FEL is required for such an experiment. Not only are high pulse energies needed, but also a very narrow probe spectrum is a prerequisite to resolve the plasmons. This was possible in our last campaign.

That way, we were able to acquire collective Thomson scattering spectra with a signal to noise ratio that does not only prove the feasibility of this experiment but would even allow single pulse exposures. This opens up the door for a time resolution of ~200 fs. Moreover, during the experiments the equipment has been constantly challenged and improved, finally leading to an outstanding performance beyond the proof of principal. This allowed us to record self-Thomson scattering spectra and their analysis is currently being carried out for a pending publication. On top of that, first experiments with the 800 nm pump and 13.5 nm FEL probe lasers have been conducted, which will also be published.

4. CONCLUSION AND OUTLOOK

Inelastic x-ray scattering (IXS) in combination with novel high peak brilliance free electron laser (FEL) light sources gives us the unique opportunity to study relaxation processes in warm dense matter with unprecedented accuracy, ultimately leading to a better knowledge of the equation of state in this regime. We have proven the feasibility of such experiments by successfully conducting them at the XUV free electron laser, FLASH. Results of self-Thomson scattering and 800 nm pump, FEL probe in warm dense hydrogen plasma and are currently being prepared for publication.

Future experiments with a variably delayed XUV pump and probe pulse are being envisaged. Those will improve accuracy as the WDM state can then be generated most homogeneously. The latest FEL sources such as LCLS and coming European XFEL bring the advantages of the FEL sources into the x-ray regime where even denser plasmas can be probed.

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